IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In the Application of:
NEIL A. ROBERTS

DUPONT CASE NO.: VK-0001 US CIP

SERIAL NO.: 10/632,817

GROUP ART UNIT: 1751

FILED: AUGUST 4, 2003

EXAMINER: JOHN R. HARDEE

FOR: REFRIGERANT COMPOSITION

DECLARATION UNDER 37 C.F.R. § 1.132
DR. DONALD B. BIVENS

I, Dr. Donald B. Bivens, declare as follows.

1. I am a retired employee from E. I. du Pont de Nemours and Company (hereinafter "DuPont"), after having worked for DuPont for 35 years on a full time basis. I am now working as a paid consultant for DuPont in the area of refrigerant development. Information about my formal education professional qualifications and experience is provided in Attachment 1, attached hereto.

2. I am providing the following data and opinions in support of the patentability of the composition consisting of (a) pentafluoroethane in an amount of from 62% to 67% by weight based on the weight of the composition;

(b) 1,1,1,2-tetrafluoroethane in an amount of from 26% to 36% by weight based on the weight of the composition, and

(c) isobutane in an amount from 3% to 4% by weight based on the weight of the composition.

My time spent in the preparation of this Declaration was paid for by DuPont. The opinions expressed herein are my own.

3. Powell, U.S. Patent No. 6,606,868

3.1 There are Too Many Possible Formulations Within the '868 Patent's Disclosure to Suggest the Compositions of Paragraph 2, above.

In order to illustrate that the '868 patent discloses a broad spectrum of possible formulations, I determined[1] the number of possible different formulations that could have been created from the '868 patent's disclosure of blends comprising:

64-76 wt.% pentafluoroethane,
24-32 wt. % 1, 1, 1, 2-tetrafluoroethane, and
0-10 wt. % of 18 different possible specifically disclosed hydrocarbons or mixtures of certain selected hydrocarbons.[2]

While refrigerant formulations are often determined and specified in fractional weight % of 0.1 %, I used an increment of 0.3 weight % for each of three components to determine the number of possible different formulations disclosed and to keep the total within calculation limit of the Minitab® statistical software. With the requirement that each formulations must add to 100 weight %, I determined that over 25,000 different

[1] This determination was made using statistical calculation software that is commercially available and marketed under the name Minitab®, a registered trademark of Minitab, Inc.

[2] The explicit teachings of the '868 patent at column 3, lines 38 -47 and the Example 7, and the claims, are to use certain hydrocarbons individually and that only a few can be used to create mixtures. The '868 patent recites the following 18 possible hydrocarbon additives: 2-methylpropane; 2, 2 – dimethyl propane, butane, pentane, 2-methylbutane, cyclopentane, hexane, 2-methylpentane, 3-methylpentane, 2,2-dimethylbutane, methylcyclopentane; and seven mixtures of hydrocarbons: n-pentane + cyclopentane + iso-pentane; and n-pentane + iso-pentane; pentane + butane; cyclopentane + butane; n-pentane + cyclopentane; iso-pentane + cyclopentane; and butane + isopentane.

formulations of two hydrofluorocarbons (R125 and R134a) and 18 specific hydrocarbon additives are possible.

I made additional calculations using an increment of 0.5 weight % for each of the three types of components, limiting to those combinations that added up to 100 weight %, and determined that it was possible to formulate over 5,000 different compositions. Both numbers (whether 25,000 or 5,000) are too large to provide any meaningful teaching or suggestion of any specific refrigerant compositions, especially one using isobutane, R125 and R134a as set forth in paragraph 2, above.

For contrast purposes, if the formulations of paragraph 2 compositions (set forth above) were limited to the 3 components that total 100%, and using a 0.3 wt. % increment, the total number of possible different formulations is only 98; and if 0.5 wt % increment, 33.

3.2 The '868 Patent Teaches Compositions that can become Flammable

An additional requirement essential to a successful formulation of an R22 alternative is nonflammability in both the as-formulated compositions and in any changed composition should there be vapor leakage of refrigerant from a storage container or refrigeration/air conditioning equipment.[3]

To determine the flammability of the '868 patent compositions after various amounts of vapor leakage, I requested two tests, (1) vapor leakage and (2) flammability to be performed on 9 different compositions selected from several of the '868 examples

[3] Flammability of compositions after specified amounts of vapor leakage is used to determine an industry recognized safety classification.

and other parts of the '868 patent – all of which use 2-4% hydrocarbon[4] content, wherein such hydrocarbon was either is n- pentane, cyclopentane, or isopentane alone or mixtures of n-butane and n-pentane.[5]

Page 2 of Exhibit A provides the original compositional data for each of the nine '868 compositions (in both liquid and vapor phases) and also for each Sample as the composition (in both liquid and vapor) changes as the vapor is permitted to leak. *See,* columns named: Original wt.%; after 50% leak; after 70% leak; after 90% leak. For example, for Sample 8, the original liquid composition was 66.1/30.7/ 3.2 wt. % of R125/R134a/n-pentane. After the Sample 8 experiences a 70% weight loss due to the vapor leak, the liquid composition changed to 49.8/43.1/7.1 wt. % of R125/R134a/n-pentane, and the 70% leak liquid composition was flammable.

Note that after performing the flammability tests for Samples Samples 1, 3, 4 and 9 which were found to be flammable having Lower Flammability Limits (LFL) and Upper Flammability Limits (UFL), flammability tests were not performed for Samples 2, 5, 6, 7 and 8. This was because, once the flammability tests for Samples 1, 3, 4 and 9 were completed, I was able to determine that each of the other five '868 patent Samples would also be flammable after some amounts of vapor leakage, as noted in the Exhibit A, page 2, last two columns "flame test results". Thus, the additional and unnecessary expense was avoided.

The resulting data (Exhibit A) show that the '868 patent compositions tested were determined to become flammable under vapor leakage conditions as prescribed by

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[4] Pentane, Isopentane, cyclopentane and two samples having both n-butane and n-pentane.

[5] These tests were performed at DuPont laboratories by personnel who run the same tests for DuPont formul ated compositions.

ASHRAE Standard 34-2004, addendum p.[6]

Samples Nos. 1-4 became flammable after a 50% weight total loss due to vapor leakage. Samples 5-6, became flammable after a 90% weight total loss due to vapor leakage. Samples 7-9, became flammable after a 70% weight total loss due to vapor leakage.

I conclude that if submitted for ASHRAE classification, the compositions of Exhibit A would be expected to receive a safety classification of flammable. This classification would result in significant use restrictions and would not be considered a favorable refrigerant for use in applications associated with the general public, such as in homes, commercial buildings, and transportation.

4.0 JP 2002-228,307 (the "JP reference"): the Mixing Procedure Reference.

4.1 In addition to discussing a method of mixing, the JP reference discusses a large number of possible compositions. In fact, the disclosure is so broad that the large number of possible formulations teaches nothing about selecting those that would be advantageous. To illustrate this point, I determined the number of possible formulations that could have been created from the JP reference's disclosure of blends comprising the recitation summarized by the Examiner, which is

55-80 weight % pentafluoroethane,

20-40 weight % 1, 1, 1, 2 – tetrafluoroethane, and

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[6] All flammability testing was done in accordance with ASHRAE Standard 34-2004, addendum p and ASTM E681, and this is the test the industry accepted test method for classifying the flammability of a refrigerant.

1-9 weight % isobutane.

Using an increment of 0.3 wt. %, and limiting the possible formulations to those that add up to 100%, the number of different possible formulations is over 4,000;[7] using a 0.5 wt. %, the number of possible formulations is over 1300. For contrast purposes, if the formulations of paragraph 2 compositions (set forth above) were limited to the 3 components that total 100%, and using a 0.3 wt. % increment, the total number of possible different formulations is only 98; and using 0.5 wt. %, the number is 33 formulations.

5.0 Unexpected Advantageous Balance of Performance and Safety is Achieved by Paragraph 2 Compositions

5.1 Despite the global search for non-ozone depleting replacements for R22, suitable replacements remain elusive. As explained following, the compositions as described in paragraph 2 above represent a discovery of refrigerant blends that provide for successful R22 replacement and achieves unexpected advantages. In particular, the compositions of paragraph 2 provide a favorable balance of performance factors such as operating within the limits of equipment designed for R22 pressures and temperatures, unexpected energy efficiency and cooling capacity near that of R22, and non-flammability safety under all leakage scenarios as required by ASHRAE Standard 34-2004, addendum p. *See* Exhibit B. Moreover, the pending application 10/632,817 further notes at [0026] that the compositions are highly compatible with mineral oil lubricants, which is a property desired by the industry.

[7] Using the Minitab® software, described *supra.*

5.2 Exhibit B contains a summary of thermodynamic refrigeration cycle calculations and refrigerant vapor leakage scenarios for the compositions of paragraph 2, above. A flame boundary plot of experimental data for mixtures of R125, R134a, and C3 – C5 hydrocarbons is also provided.[8] From these data, one learns that over the range of the compositions of paragraph 2, the compositions achieve similar cooling capacity and energy efficiency as R22, and are non flammable under all leakage scenarios as required for evaluation by ASHRAE Standard 34-2004, addendum p.

5.3 In addition, the compositions of paragraph 2 have a zero-ozone depletion potential ("ODP")[9] and as can be seen from the calculated data provided in Exhibit B, using the Cycle D thermodynamic cycle calculation software program available from the U.S. National Institute of Standards and Technology, the paragraph 2 compositions exhibit the following additional favorable performance factors:

low temperature glide;
suction pressure nearly equal to that of R22;
lower discharge temperature than that of R22;
energy efficiency rating that is substantially equal to that of R22;
cooling capacity within 6% of that of R22; and
slightly higher discharge pressure than that of R22, but within the system design limits.

While Cycle D is a useful thermodynamic cycle calculation program, it does have

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[8] The Flame Boundary Plot, of Exhibits C and D, was collected by DuPont over many years, including experiments performed and data collected after 2003. This chart was not made available to the public by DuPont until about June 2006.

[9] The ODP or Ozone Depletion Potential is measured for a single molecule of the refrigerant to destroy Ozone. All refrigerants use R-11 as a datum reference where R11 has an ODP of 1.0 compared with R22's ODP of 0.040.

limitations. For example, it does not provide the complete information needed to formulate a refrigerant with an optimum balance of safety and performance. Moreover, to the extent the thermodynamic cycle data that it provides is useful, the cycle data are determined only after a refrigerant formulation has been selected and its formulation entered into the software. Cycle D is not useful in identifying possible recipes of refrigerant formulations, (from the thousands of possible components) to meet a pre-selected specified performance criteria. Rather, it is used solely to provide Cycle data for the "pre-selected" recipe. Once the Cycle data is obtained, further research and development is performed.

5.4 Actual system test data provided in the 10/632,817 ('817 application) is consistent with the favorable performance of the thermodynamic cycle calculations obtained from the Cycle D data provided in Exhibit B.

In particular, for one of the formulations of paragraph 2, the '817 application has performance data for testing the formulation in a compressor calorimeter system, as compared with that of R22. *See*, pages 4-5 of the '817 application, Tables 1, 2, and 4. The compressor calorimeter system is described on page 3 of the '817 application, section [0039] to [0044]. That system is described as having a compressor, oil separator, condenser, automatic expansion valve, and an evaporator, which allows for system design effects to be a factor in performance of the formulations of paragraph 2 versus that of R22. The system test data clearly show the "isobutane blend" of R125/R134a/R600a (64.9%/31.7%/3.4%) had favorable performance as a replacement for R22. *See also* attached as Exhibit D, a graphical representation of the data of Table 2 (R22) and Table 4 ("isobutane blend"). In particular, the "isobutane blend" had an evaporator pressure

within 5% of R22 (1.4 psia higher at 30 psia), 40 degrees C lower discharge temperature (which means lower thermal degradation effects on the system and the refrigerant/lubricant), energy efficiency of about 99% of R22, cooling capacity of about 94% of R22, and compressor discharge pressure of about 7% higher than R22. These data show the "isobutane blend" and the other formulations of paragraph 2 can be successfully used in existing R22 systems with minimal changes to the equipment and operating conditions.

5.5 The conclusion from using the Cycle D data (as supported by actual system data), flammability testing and analysis, is that the compositions of paragraph 2 have the unexpected and advantageous balance of properties over the full range of compositions.

CONCLUSION

I conclude that the compositions as described in paragraph 2 are, unexpectedly, good replacements for R22 over the full range of possible formulations. I further conclude that the non-flammable formulations possess the unexpected and advantageous balance of the variety of performance requirements and operational factors that are difficult to find in non-ozone depleting refrigerants.

I declare under penalty of perjury that the above facts are true to the best of my knowledge.

Signed this 21st day of March, 2007,

Dr. Donald B. Bivens

Dr. Donald B. Bivens

Sworn to and subscribed before me this 21st day of March, 2007,

Signature: Barbara J. Massie

Notary Public Name: Barbara J. Massie

My Commission Expires: July 28 2007 (Notary Seal)

BARBARA J. MASSIE
NOTARY PUBLIC
STATE OF DELAWARE
My Commission Expires July 28, 2007

LIST OF EXHIBITS

EXHIBIT A -- Vapor Leak and Flammability Test Results for '868 Patent Formulations

EXHIBIT B -- Cycle D Performance; Vapor Leak Test, and Flame Boundary Diagram.

EXHIBIT C-- Butane means n-butane.

EXHIBIT D-- '817 Application Graphical Representation of Tables 2 and 4.